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SUPPORTING DOCUMENT

1 Page 1 of 29

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6 Author

J. E. Cummings
Name (Type or Print)

J. E. Cummings
Signature

80423
Organization Code

7 Abstract

This report is provided to support an engineering study being prepared on alternative methods to sample, analyze, and decommission Tank 241-CX-72 located at the Strontium Semiworks complex. An investigation of the available historical documentation was conducted to provide information on the waste characteristics of the contents of this tank. This report also incorporates radiological characterization investigations recently conducted.

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TANK 241-CX-72 PRELIMINARY WASTE CHARACTERIZATION

June 30, 1989

Prepared by: J. E. Cummings 6/30/89
J. E. Cummings, Senior Engineer Date
Decommissioning Engineering
Environmental Division

Reviewed by: V. Subrahmanyam 6/30/89
V. B. Subrahmanyam Date
Analytical Systems Laboratories
Chemical Processing Division

Westinghouse Hanford Company
Post Office Box 1970
Richland, Washington 99352

TANK 241-CX-72 PRELIMINARY WASTE CHARACTERIZATION

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TANK 241-CX-72 PRELIMINARY CHARACTERIZATION

EXECUTIVE SUMMARY

This report is provided to support an engineering study being prepared on alternative methods to sample, analyze, and decommission the 241-CX-72 tank located at the Strontium Semiworks complex. An investigation of the available historical documentation on this tank was conducted to provide some information on the possible radiological and chemical constituents in this tank. This investigation includes review of available documents, search of laboratory records for evidence of previous sampling and analyses, and discussions with present as well as retired employees who are familiar with the history of this tank.

The Strontium Semiworks records were sent to the Record Center after the Reduction Oxidation (REDOX) and Plutonium Uranium Extraction (PUREX) processes. Most of these records were destroyed in 1982. No microfiche of these records exist. Some early documentation does exist at the Battelle Library, some of which is classified and therefore access is limited. An extensive review of the historical documents that were available has revealed a number of inconsistencies. This is due in part to the nature of record keeping and data collection methods used during the 1950s and 1960s.

Tank 241-CX-72 is a cylindrical, single-shell, carbon steel tank, approximately 40-in. in diameter and approximately 36 ft long. The tank is set inside of a caisson and is situated vertically approximately 14 ft below grade. A sketch of the configuration of Tank 241-CX-72 is attached. The time sequence of historical events at Strontium Semiworks is also attached.

Based on available historical documentation, Tank 241-CX-72 was used as an experimental tank to determine the characteristics of self-concentrating wastes. The tank received waste from the PUREX process. The PUREX process used tributyl phosphate in kerosene solvent to extract plutonium and uranium from acid solutions of irradiated uranium. Nitric acid was used to promote extraction of plutonium and uranium. Process condensates from the PUREX process contained predominantly dilute nitric acid and other inorganic contaminants. Phenomena of waste self-concentration and "bumping" in tanks by internally-generated, externally-introduced energy was also investigated.

Based on personal contacts with now-retired personnel once associated with Strontium Semiworks, there exists the possibility that the decontamination flushes following the PUREX processes were also routed to this tank. A list of the decontamination solutions used for the various contamination and fission product species is attached.

Tank 241-CX-72 was not directly associated with any disposal cribs or any other tanks. Speculation regarding the constituents of Tank 241-CX-72 based on the known constituents of Tank 241-CX-70 is beyond the scope of this report. However, although the extraction efficiencies of the REDOX and PUREX processes were different, the waste from processing irradiated material would be expected to contain the same species of radionuclides.

EXECUTIVE SUMMARY (Cont'd)

A formal waste designation has not been performed on the waste in this tank because the quantity, concentrations, and current chemical compositions are not known. It is uncertain whether the decontamination flushes used after the PUREX process were routed to Tank 241-CX-72. No documentation regarding this has been located; however, personal contacts with personnel familiar with the Strontium Semiworks complex recall that the intent was to store the decontamination flushes in this tank. Although the composition of the decontamination flushes used is known, the quantity used for each flush is not known.

If the decontamination flushes did indeed go to this tank, then the presence of chromium would cause the tank contents to be designated as potentially DANGEROUS WASTE. Other constituents, especially fluoride, would also influence the DANGEROUS WASTE designation.

Recent radiological characterization investigations were conducted by Dr. Vishnu B. Subrahmanyam of Analytical Systems Laboratories and are summarized as follows:

- Radiation dose rate and neutron flux measurements strongly suggest the presence of a minimum 10 ft layer of radioactivity at the tank bottom. The temperature readings in the drywell support the presence of an internal energy source.

- A close examination of radiation dose rates at different depths revealed a step-wise increase (approximately ten fold) in 5-ft intervals. This is most likely a result of the past practice of maintaining as high a liquid level as practical in self-concentrating tanks.

- Gamma spectroscopic investigation with a shielded detector equipped with a window provided evidence for the presence of radioactive material in the tank. The gamma spectrum at 19-ft depth contained only the gamma ray of Cs-137. Higher activity levels and possibly other gamma emitters are present at deeper locations.

- Irradiation of indium foils at a depth of 47 ft confirmed the presence of essentially fast neutrons.

- The activity layer is dry and contains little, if any, hydrogenous materials to thermalize the neutrons generated within the contents of the tank.

- Based on an objective assessment and evaluation of all pertinent data, it is concluded that the activity layer at the bottom of Tank 241-CX-72 contains fission product activities mixed with a most probable 150 to 200 grams of the fissile isotope, Pu-239.

1.0 INTRODUCTION

This report is provided to support an engineering study being prepared on alternative methods to sample, analyze, and decommission the 241-CX-72 tank located at the Strontium Semiworks at the 200-E area. An investigation of the available historical documentation on this tank was conducted to provide some information on the possible radiological and chemical constituents in this tank. This investigation includes review of available documents, search of laboratory records for evidence of previous sampling and analyses, and discussions with present as well as retired employees who are familiar with the history of this tank.

Based on historical documentation, Tank 241-CX-72 was used as an experimental tank to determine the characteristics of self-concentrating wastes. It was used during the PUREX operations. Based on personal contacts with employees (now retired) who are familiar with the history of the Strontium Semiworks complex, decontamination flushes following the termination of the PUREX operations may also have been routed to Tank 241-CX-72.

Tank 241-CX-72 was not directly associated with any cribs or any other tanks. Although speculation regarding the constituents of Tank 241-CX-72 based on the known constituents of Tank 241-CX-70 is beyond the scope of this study, information on Tank 241-CX-70 is presented in this report for information purposes.

This report also incorporates recent radiological characterization investigations performed by Dr. Vishnu B. Subrahmanyam of Analytical Systems Laboratories. Dr. Subrahmanyam expects to have formal, more detailed documentation of his studies issued in July 1989.

The time sequence of events associated with the Strontium Semiworks complex is shown in Attachment 1. The configuration of Tank 241-CX-72 is shown in Attachment 2.

2.0 HISTORICAL DOCUMENTATION

Because of the great length of time that this tank has been in existence and the nature of historical record keeping practices and resultant conflicting information, the historical data presented here is coordinated directly with the corresponding reference source. Quotation marks have been eliminated.

2.1 HISTORY OF STRONTIUM SEMIWORKS AND TIME SEQUENCE OF EVENTS

NOTE: See Attachment 1 for the time sequence of historical events.

2.1.1 Ref: "Level I Remedial Investigation Work Plan - 200 Area Strontium Semiworks Liquid Waste Disposal Sites", prepared by Pacific Northwest Laboratories (PNL), issued September 1987

PAGE 2-25: The REDOX process was the first process to recover both plutonium and uranium from irradiated fuel materials. The process used a solvent extraction process to remove plutonium and

2.0 HISTORICAL DOCUMENTATION (Cont'd)

2.1 HISTORY OF STRONTIUM SEMIWORKS AND TIME SEQUENCE OF EVENTS (Cont'd)

uranium from dissolved fuels into a methyl isobutyl ketone (hexone) solvent. Waste streams from the REDOX process were slightly acidic and contained fission products and large volumes of aluminum nitrate used to promote extraction of the plutonium and uranium.

PAGE 2-25: The Strontium Semiworks facility was placed in service in 1952 and was first used in demonstration of the REDuction and OXidation (REDOX) process and later for demonstration of the Plutonium and Uranium Recovery through EXtraction (PUREX) process until 1956. The facility was out of service from 1956 to 1960 and was then modified for recovery and purification of Sr-90 (Strontium 90) from other Hanford reprocessing plant by-products. The Semiworks plant was retired in 1967.

PAGE 2-25: The Strontium Recovery Process performed at the Semiworks facility utilized a complex liquid organic ion exchanger, di-2-ethyl-hexyl phosphoric acid, to extract strontium from acid solutions of waste fuels.

2.2 PURPOSE OF TANK 241-CX-72

2.2.1 Ref: Memo, Harlow/Teal, "Disposition and Isolation of Tanks 270-E-1, 270-W, 241-CX-70, 241-CX-71, and 241-CX-72", dated July 2, 1974.

PAGE 4: Tank 241-CX-72 was used as an experimental tank to determine the characteristics of self-concentrating wastes during 1956.

2.2.2 Ref: SD-DD-FL-001, Rev 0.0, "Rockwell Retired Contaminated Facility Listing and Description", by A. A. Crusselle and T. Romano, dated July 1982.

PAGE 70: The 241-CX-72 waste self concentrator was an experimental project operated in the 1950s by Hanford Labs. It was used for PUREX waste, which was then piped to 241-C Tank Farm. It has not been used since, and is considered retired.

2.3 DESCRIPTION OF TANK 241-CX-72 AND QUANTITY OF CONTENTS

See Attachment 2, Tank 241-CX-72 Configuration.

Location: 200 East Area - N41900/W50100

Reference Drawings: H-2-4422

2.0 HISTORICAL DOCUMENTATION (Cont'd)

2.3 DESCRIPTION OF TANK 241-CX-72 AND QUANTITY OF CONTENTS (Cont'd)

2.3.1 Ref: SD-DD-FL-001, Rev 0.0, "Rockwell Retired Contaminated Facility Listing and Description", by A. A. Crusselle and T. Romano, dated July 1982.

PAGE 70: The 241-CX-72 tank is set inside a caisson which is a carbon steel cylinder 6 ft-1 in. in diameter and 36 ft-6 in. in length, buried upright approximately 14 ft below grade. The tank is approximately 4 ft in diameter and 35 ft-8 in. in length and is an agitator tank. Several risers extend from the top of the tank, and are visible above grade.

2.3.2 Ref: Harlow/Teal, "Disposition and Isolation of Tanks 270-E-1, 270-W, 241-CX-70, 241-CX-71, and 241-CX-72", dated July 2, 1974.

PAGE 4: Tank 241-CX-72 is a 36 ft deep by 3 ft in diameter carbon steel tank located just east of tank 241-CX-70. The only inlet to this tank is from the 201-C Building, and it is cut and capped there. There is no exit from the tank, but there is an above-ground vent riser. Currently, liquid level measurements are six feet, two-and-one-half inches of which all but approximately one inch is sludge. This level is measured daily and is holding steady.

2.3.3 Ref: SD-WM-SAR-003 issued, dated March 1984.

Page 5-49: Tank CX-72 is an upright cylindrical vessel 4 ft in diameter and 35 ft-8 in. in length. It rests on the caisson concrete pad with support pads welded to the tank bottom. The 3/8-in. thick vessel walls are reinforced with five stiffener rings that extend nearly out to the caisson wall. Three rows of vertical guides connect the stiffener rings. A cylindrical electric heater is mounted just above each stiffener ring. The top of the vessel is sealed with a plate that also extends out over the caisson and seals the caisson. Five pipes extend from the tank to above grade level, and two pipelines enter the tank underground from the vault. A manually operated agitator extends from the vessel to above ground.

PAGE 5-56: Tank 241-CX-72 is set inside a caisson which is a cylinder fabricated from 1/2-in. carbon steel plate. The caisson is 6 ft in diameter and 36 ft-6 in. long, buried upright approximately 14 ft below grade. The bottom of the caisson is sealed with a 12-in. thick reinforced grout plug that provides a base pad for Tank 241-CX-72.

2.0 HISTORICAL DOCUMENTATION (Cont'd)

2.3 DESCRIPTION OF TANK 241-CX-72 AND QUANTITY OF CONTENTS (Cont'd)

2.3.4 Ref: Memo, Mirabella/Dukelow, "Aux Tanks, Sumps, and Vaults Solids and Liquid Volumes", dated March 3, 1978.

Identification: CX-72
Capacity (Gal): 20,000
Liquid (Gal): 0
Solid (Gal): 0
Comments: Tank is empty.

2.3.5 Ref: Aux. Tanks - Approx. Stabil. Dates, dated August 1978.

Capacity (K gal): 20.0
Liquid (K gal): 0
Solids (K gal): 0
Total (K gal): 0
Requires Sampling: No
Active/Inactive: Inactive
Comments: Tank is empty.

2.3.6 Ref: Aux. Tanks - Approx. Stabil. Dates, dated August 25, 1978.

Capacity (K gal): 20.0
Liquid (K gal): 0
Solids (K gal): 0
Total (K gal): 0
Liquids Level (inches): ---
Solids Level (inches): ---
Requires Sampling: No
Active/Inactive: Inactive
Comments: Tank is empty.
Status: Tank is stabilized.
Approx. Stab. Date: Primary Stab.

2.3.7 Ref: Aux. Tanks - Stabil. Plan, dated January 11, 1979.

Capacity (K gal): 20.0
Liquid (K gal): 0
Drainable Liquid (K gal): 0
Solids (K gal): 0
Total (K gal): 0
Liquids Level (inches): ---
Solids Level (inches): ---
Sampling: EF
Active/Inactive: Inactive
Comments: Tank is empty.
Approx. Stab. Date: Interim Stab.
Stabilization Plan: INTERIM STABILIZED

2.0 HISTORICAL DOCUMENTATION (Cont'd)

2.3 DESCRIPTION OF TANK 241-CX-72 AND QUANTITY OF CONTENTS (Cont'd)

2.3.8 Ref: HW52860, "Standby Status Report - Hot Semiworks Facility" by C. R. Cooley, dated September 1, 1957.

Total Volume Received in Tk-72: 8,723.90 Liters
Pounds U: 61.22
Grams Pu: 48.49

2.4 HISTORICAL TANK RADIONUCLIDE DOCUMENTATION

2.4.1 Ref: Memo, Harlow/Teal, "Disposition and Isolation of Tanks 270-E-1, 270-W, 241-CX-70, 241-CX-71, and 241-CX-72" dated July 2, 1974.

PAGE 5: Analysis of the solution shows a clear, light brown liquid with a pH of 9.50 and a trace of solids. The over-the-top radiation reading is less than 0.001 mR per hour. Other results are:

Pu: 1.12 x 10(-8) g/gal
U: 2.43 x 10(-3) g/gal
Cx-137: None detected
Sr-89, 90: 4.33 x 10(-3) microcuries/gal
DTA: No exotherms

2.4.2 Ref: DOI05ER00001, Rev 00 issued, "Preliminary Study for Decontamination and Decommissioning of Strontium Semiworks" by D. E. Bowers, dated November 13, 1979.

PAGE 15: Tank 72 and vault have trace transuranic (TRU), fission product, uranium, and thorium contamination; no estimates are available. It is believed that Tank 72 may actually be moderately or grossly contaminated with fission products.

2.4.3 Ref: SD-DD-ES-003 issued, "Strontium Semiworks Decommissioning Engineering Study" by J. M. Marzec, dated October 27, 1983.

PAGE 123: CX-72 Radiation Data*: Pu=3 Ci, Beta=6000 Ci

*Inventories are preliminary estimates made from knowledge of the operations and history of the facilities.

2.4.4 Ref: SD-WM-SAR-003 issued, dated March 1984.

PAGE 5-56: The 241-CX-72 Tank and Vault contain low levels of residual contamination. The 20,000-gal tank and the caisson annular space are empty. Specific data on vault contamination levels are not available, but are estimated to be 3 Ci TRU and 6000 Ci beta.

2.0 HISTORICAL DOCUMENTATION (Cont'd)

2.4 HISTORICAL TANK RADIONUCLIDE DOCUMENTATION (Cont'd)

2.4.5 Ref: SD-DD-FL-001, Rev 0.0, "Rockwell Retired Contaminated Facility Listing and Description", by A. A. Crusselle and T. Romano, dated July 1982.

PAGE 71: Radionuclide Inventory: 3 curies Pu, 6,000 curies beta.

2.5 HISTORICAL TANK CHEMICAL DOCUMENTATION

2.5.1 Ref: "Level I Remedial Investigation Work Plan 0 200 Area Strontium Semiworks Liquid Waste Disposal Sites", prepared by PNL, issued September 1987.

NOTE: Historical documentation indicates that Tank CX-72 was operated during the PUREX phase only. However, Tank CX-72 may have also been used to store the decontamination flushes which occurred following termination of the PUREX operations.

PAGE 2-25: The PUREX process is an advanced extraction process using tributyl phosphate in kerosene solvent to extract plutonium and uranium from acid solutions of irradiated uranium. Nitric acid is used to promote extraction of plutonium and uranium as opposed to the metallic nitrates used in the REDOX process. Process condensates from the PUREX Process contain predominantly dilute nitric acid and other inorganic contaminants. The volume of high level waste produced by the PUREX process is much lower per unit of fuel processed than those of previously applied processes.

2.5.2 Ref: HW-52860, Undocumented, "Standby Status Report - Hot Semiworks Facility, by C. R. Cooley, dated September 1, 1957.

NOTE: Based on personal contacts with employees familiar with the history of the Strontium Semiworks, decontamination flushes following termination of the PUREX operations may also have been routed to Tank 241-CX-72.

See Attachment 3, Decontamination Solutions.

PAGE 46: Some decontaminating reagents were determined by sending actual samples of the material to be decontaminated to the laboratory and subjecting it to a series of solutions to find which one was the most effective. Many times, the choice of the agent used was quite arbitrary since it was not known what particular species of fission product or combination of products were present. Generally, however, the principal contenders were zirconium and niobium accompanied by some ruthenium. CT (caustic-tartrate, "hot")

2.0 HISTORICAL DOCUMENTATION (Cont'd)

2.5 HISTORICAL TANK CHEMICAL DOCUMENTATION (Cont'd)

was used quite extensively for flushing. This was generally followed with "hot" oxalic acid. Oxalic acid was generally "killed" with hydrogen peroxide to reduce waste volume and to allow more efficient concentration (concentration of oxalic solutions is difficult) (1/7 volume of 30 percent H_2O_2 is required to kill one volume of 10 percent oxalic acid. Seventy percent is added cold and the remainder after the solution is heated to boiling).

PAGE 46: Another combination which seemed to be effective against ruthenium was PC (permanganate-caustic). This was generally followed with N-FAS (nitric ferrous ammonium sulfate) to remove the precipitated manganese dioxide. A 3-20 (fluoride nitric) flush was generally used at least once during the decontamination campaign for final cleanup. Its repeated use is not recommended because of its corrosive nature. For concrete and painted surfaces, the above flushes were generally followed with an ND (nitric dichromate) flush to remove cesium and cerium.

PAGE 46: A flow system was set up to decontaminate the tanks and the solution was transferred progressively from one tank to another. The transfers were made from low to a high contamination tank. All tanks were generally filled to near overflowing to provide solution contact with the tank top. During the course of these tank transfers, all of the auxiliary lines to each tank were flushed as well.

2.6 HISTORICAL CHEMICAL/RADIOLOGICAL DOCUMENTATION OF ASSOCIATED TANKS/CRIBS

NOTE: Tank 241-CX-72 was not directly associated with any cribs or any other tanks. Speculation regarding the constituents of Tank 241-CX-72, based on the known constituents of Tank 24-CX-70, is beyond the scope of this report. The following documentation regarding Tank 241-CX-70 is presented for informational purposes.

2.6.1 Ref: Memo, M. T. Jansky/G. E. Entrop, "Analysis of Tank CX-70 Waste, dated December 10, 1985.

See Attachment 4 for the chemical and radionuclide composition of Tank 241-CX-70.

NOTE: The data for this reference was obtained from samples taken prior to initiation of waste removal. The initial grab sample, collected by bottle and string, represented the tank sludge inventory of 10,300 gallons at that time. Because of the sampling method utilized and the two-phase characteristic (solid and liquid) of the sludge, accuracy of the sampling is highly questionable.

2.0 HISTORICAL DOCUMENTATION (Cont'd)

2.7 HISTORICAL SAMPLING

2.7.1 Ref: Letter, Malody to Elgert of Energy Research and Development Administration (ERDA), "Waste Tank Survey" dated November 1976.

Sampling was attempted in another location in the tank but no sludge was found. Sludge measurements and visual inspection of the tank indicate that there is no sludge in the tank. Optical equipment is being purchased which will allow us to obtain an in-tank view of tank CX-72.

2.7.2 Ref: Telephone Conversation, J. E. Cummings/Dave Briggs (222-S Laboratory).

On May 5, 1989, the 222-S Laboratory was requested to perform a computer and hard copy record search for evidence of any sampling done on this tank. No records of sampling were located.

3.0 RECENT RADIOLOGICAL INVESTIGATIONS

The following investigations were recently conducted by Dr. Vishnu B. Subrahmanyam of Analytical Systems Laboratories. Most investigations were conducted inside of the 3-in. drywell.

- Gamma spectroscopic investigation with a shielded detector equipped with a window provided evidence for the presence of radioactive material in the tank. The gamma spectrum at 19 ft depth in the drywell showed the presence of only Cs-137. Maximum activity of this isotope was observed with the collimator window directed towards the center axis of the tank. A 40% lower value was detected when the window was rotated by 180 degrees. Higher activity levels and possibly other gamma emitters are present at deeper locations.

- The results of relative axial profiles of neutron flux and dose rate measurements are shown in Attachment 5. Radiation dose rate and neutron flux measurements strongly suggest the presence of a minimum 10 feet layer of radioactivity at the tank bottom. The temperature readings in the drywell also support the presence of an internal energy source.

- Irradiation of indium foils at a depth of 47 ft confirmed the presence of essentially fast neutrons. Measurements at depths below 40 ft with a thermal neutron detector ($^{10}BF_3$ tube) wrapped with 0.5 inch polyethylene, 0.5 inch lead, or 0.5 inch lead with an additional 1/16 cadmium foil showed that greater than 90% of the neutrons have energies above the cadmium cut off energy. The absence of any thermal moderation of neutrons can be attributed to the reported experimental determination of 2% liquid in waste when its volume was reduced by a factor of five.

3.0 RECENT RADIOLOGICAL INVESTIGATIONS (Cont'd)

- The neutron flux determined from indium foil irradiations combined with the geometry of irradiation resulted in a calculated value of 7 neutrons per second per cubic centimeter of a cylindrical source with uniformly distributed neutron sources.

- The gamma spectrum of indium foil exposed overnight to neutrons at the 47 ft depth contained only the 341 keV gamma ray attributable to the (n,n') reaction with a cross-section of 174 mb averaged over the energy spectrum of fission neutrons. Intense gamma rays expected from the thermal neutron capture product were not observed in spite of the 157 b formation cross-section. Epithermal energies of the neutrons at this location were confirmed by count rates of a thermal neutron detector wrapped with different shielding materials (see above).

- A neutron flux value of 141 neutrons per sq cm per second was calculated from the measured activity of the (n,n') reaction product and the irradiation parameters. Taking into consideration the irradiation geometry, verified by numerical integration, the measured flux corresponds to a bulk neutron generation rate of 7 neutrons per second per cubic centimeter.

- Concentration of transuranium isotopes consistent with the calculated bulk neutron production rate depends on the relative isotopic composition and, more importantly, on their chemical form of existence. Three different isotopic compositions (weapons grade plutonium, dissolved fuel irradiated to 9% of Pu-240, and the composition based on smear samples obtained from a deep bed fiberglass filter) and two chemical forms (oxides and fluorides) were considered.

- Calculated values of Pu-239 present as oxide in an assumed 11 ft layer of activity with the three isotopic compositions were, respectively: 149, 51, and 0.65 kg. The corresponding values for fluoride forms were 2.2, 0.80, and 0.16 kg. Based on the source of the waste stored in Tank CX-72 (pilot studies of PUREX process) with possible addition of other fluids with fluoride ions, the transuranics are most likely present as fluorides.

- The estimates of 51 or 0.65 kg of dissolved fuel in the tank is unrealistic since only the unrecovered plutonium and other transplutonium isotopes of the processed fuel are most likely to be left in the waste sent to the tank. Again, assuming 2% of the plutonium content of the processed 9% burn up fuel as being present in the tank waste, the calculated fissile content values were 807 and 290 grams present respectively as oxide and fluoride.

3.0 RECENT RADIOLOGICAL INVESTIGATIONS (Cont'd)

- A close examination of radiation dose rates at different depths revealed a step-wise increase (approximately ten fold) in 5-ft intervals. This is most likely a result of the practice at the time the tank was used to maintain as high a liquid level as practical in the self-concentrating tanks.

- It was learned from B. J. Saueressig that the tanks 70, 71, and 72 did not receive any waste from the strontium recovery operations also conducted in the facility.

- It was reported by R. E. Tomlinson, one of the engineers engaged in development work at Hot Semiworks that the contents of tank CX-72 consists of the liquid high level waste (HLW) from the pilot scale studies of the PUREX process. This tank was also reported to have been used to investigate the "bumping" associated with wastes with internal energy sources or when the energy is provided from an external source.

- It was also learned from conversations with Mr. Tomlinson that CX-72 tank received liquid waste from "heel" recovery studies as well as from zirconium cladding dissolution investigations, processes that most likely utilized fluorides and/or fluorinated solvents. The possible discharge of solutions containing fluoride ion used for decontaminating the operational areas was also mentioned.

- The information obtained in conversations with R. E. Tomlinson was for the most part corroborated by F. W. Woodfield, who was a Manager at the Hot Semiworks during the pilot studies.

4.0 DISCUSSION

Because of the great length of time that Tank 241-CX-72 has been in existence and the nature of historical record keeping practices and resultant conflicting information, the accuracy of some of the information obtained from the historical documentation is highly suspect. As an example, historical documentation repeatedly states that the capacity of Tank 241-CX-72 is 20,000 gallons, when in fact the capacity is 5,000 gallons. In addition, the various documentation on the physical description of this tank is highly inaccurate. There was a tendency over the years to assume that a documented statement was accurate and therefore the same erroneous statement was repeated in subsequent documents.

4.1 DISCUSSION OF HISTORICAL RADIOLOGICAL/CHEMICAL INFORMATION

NOTE: Tank 241-CX-72 was used during the PUREX operations and possibly the decontamination flushes following termination of the PUREX operations.

4.0 DISCUSSION (Cont'd)

4.1 DISCUSSION OF HISTORICAL RADIOLOGICAL/CHEMICAL INFORMATION (Cont'd)

The documented inventory of 3 curies of plutonium and 6,000 curies of beta, although repeated in many documents, was given as a rough estimate and cannot be corroborated.

Although no detailed historical documentation of radionuclide inventory is available for Tank 241-CX-72, there is both chemical radionuclide documentation on Tank 241-CX-70 contents. Because Tank 241-CX-72 was not directly associated with other tanks, speculation regarding the constituents of Tank 2410-CX-72, based on the known constituents of Tank 24-CX-70, is beyond the scope of this report. However, although the extraction efficiencies of the REDOX and PUREX processes were different, the waste from processing irradiated material would be expected to contain the same species of radionuclides.

The PUREX process was an advanced extraction process using tributyl phosphate in kerosene solvent to extract plutonium and uranium from acid solutions of irradiated uranium. Nitric acid was used to promote extraction of plutonium and uranium as opposed to the metallic nitrates used in the REDOX process. Process condensates from the PUREX process contained predominantly dilute nitric acid and other inorganic contaminants.

Based on historical documentation, it is known that the decontamination flushes following the PUREX operations went to Tank 241-CX-70 and Tank 241-CX-71. Mr. R. E. (Roy) Tomlinson (now retired) recalls that the intent was also to store the flush waste in Tank 241-CX-72. See Attachment 3 for a list of the decontamination solutions used following PUREX operations.

The list of decontamination solutions on Attachment 3 includes 1-14% Turco 4182A and 10-20% Oakite #31. Contact was made with Hanford Environmental Health Foundation (HEHF) to ascertain the chemical constituents of these two solutions. Turco 4182A contains Ammonium Bicarbonate (50% by weight) and Sodium Hexametaphosphate (45% by weight). Oakite is no longer used; however, HEHF has contacted the past manufacturer to obtain the chemical constituents in Oakite. Oakite #31 contains phosphoric acid (60-70%) and Nonylphenoxy Polyethoxy Ethanol (less than 5%). The information on Turco 4182A provided from HEHF is based on the current formula. A formula change over the years is possible but no record to ascertain this is available.

Although the types of chemicals and chemical compositions used were documented, the quantities used and the current compositions are unknown.

A formal waste designation has not been performed on the waste in this tank because the quantity, concentrations, and current chemical compositions are not known.

4.0 DISCUSSION (Cont'd)

4.1 DISCUSSION OF HISTORICAL RADIOLOGICAL/CHEMICAL INFORMATION (Cont'd)

It is known that Sodium Dichromate was used in the decontamination flushes. It is uncertain whether the decontamination flushes went to 241-CX-72. If the decontamination flushes did indeed go to this tank, then the presence of chromium would cause the tank to be designated as a potentially DANGEROUS WASTE. Other constituents, especially fluoride, would also influence the DANGEROUS WASTE designation.

4.2 DISCUSSION OF RECENT RADIOLOGICAL INVESTIGATIONS

Recent radiological characterization investigations were conducted by Dr. Vishnu B. Subrahmanyam of Analytical Systems Laboratories and are as follows:

- The measured 40% of higher activity of Cs-137 with the collimator window directed towards the center axis of tank CX-72 at a depth of 19 ft provides strong support for the presence of fission product activity in the tank. Because of the 75% attenuation of Cs-137 photons by the 1/2 inch lead shielding and fairly wide field of view through the collimator window, it is not possible to determine whether Cs-137 activity exists as a thin layer on the walls of the tank and/or the outer surface of the drywell. Measurements of a similar nature with a narrower view cone are required to confirm this possibility.

- The relative neutron flux and radiation dose profiles (see Attachment 5) clearly indicate the distinct boundary of an activity layer at a depth of about 40 ft. The constant levels of these two profiles also suggest the uniform distribution of activity in the contents of the tank at depths below 40 ft with likely higher concentrations in the bottom 2 to 3 ft. This activity region can be determined to be approximately 11 ft based on the record drawings of Tank CX-72 that establish the bottom of the tank at a depth of 51 ft.

- A close examination of the radiation dose readings revealed that the dose rates can be approximated by a constant value in 5 ft length segments. This stepwise approximately ten fold increase in dose rate every 5 feet is mostly a consequence of the past practice of maintaining as high a liquid level as possible in self-concentration tanks. Also, the sudden increase of activity with possible presence of other fission product gamma emitters at about 20 ft depth apparently precluded the accumulation of a gamma spectrum below 19 ft depth.

- The investigation of neutron flux measurement with indium foil irradiation at 47 ft depth resulted in the observation of only the 341 keV gamma ray of the product formed by (n,n') nuclear reaction. The detection of the product with a formation cross-section of only 174 mb and absence of expected intense gamma rays from the thermal

4.0 DISCUSSION (Cont'd)

4.2 DISCUSSION OF RECENT RADIOLOGICAL INVESTIGATIONS (Cont'd)

neutron capture product 116m Indium with a formation cross-section of 157 b, clearly suggests that the thermal neutrons constitute only a negligible fraction of the neutrons at that depth.

- The epithermal energies of neutrons at the 47 ft depth in the 3 inch diameter drywell were further supported by the investigations with a thermal neutron ($^{10}BF_3$) detector. The ratio of neutron count rates of approximately 3 at depths below 40 ft with a 0.5 inch lead shielding and with the additional 1/16 inch cadmium confirm that the energies of neutrons are above the cadmium cut-off energy of 0.5 eV. The significant increase in neutron count rate observed by replacing the lead shield by an equal thickness of polyethylene is further evidence of the epithermal energies of the neutrons. This neutron count data can be shown to be consistent with greater than 90% component of high energy neutrons.

- The observation of high energy neutrons generated in the contents of Tank CX-72 and the apparent absence of any moderating constituents can be reconciled on the basis of the information from historical records and the results of self-concentrating of waste stored in tanks. The addition of 8724 liters of liquid waste to Tank CX-72, containing 61.2 pounds of uranium and 48.49 g of plutonium, was reported in 1957. In 1974, it was reported to occupy 74.5 inches with only 1 inch of liquid. This is equivalent to a reduction in volume in excess of a factor of 5. This observation coupled with the experimental data from waste concentration studies that reported less than 2% liquid content when the waste volume was reduced by a factor of 5 and the lapse of time since 1974, strongly indicate that the sediment layer at the bottom of tank CX-72 is essentially free of any liquid. Hence, the absence of any neutron energy moderating constituents.

- The indium foil irradiation data allow for the evaluation of an epithermal neutron flux of 141 neutrons per second per square centimeter. Based on the geometry of irradiation and utilizing a reasonable relaxation length of 25 cm, this flux was shown to be 20 times the specific bulk neutron production rate (neutrons per second per cubic centimeter). This conversion factor was also confirmed by a numerical integration method. It is therefore concluded that the seven neutrons per second are generated per cubic centimeter of the 40 inch diameter cylindrical source with uniformly distributed neutron sources.

- The neutrons are generated in the 11 ft activity layer by spontaneous fissions and by (alpha,neutron) reactions. Therefore, any attempt to calculate the bulk concentration of transuranium isotopes requires not only a knowledge of the transuranic (TRU) isotopic composition but also the chemical form of these isotopes. The latter chemical

4.0 DISCUSSION (Cont'd)

4.2 DISCUSSION OF RECENT RADIOLOGICAL INVESTIGATIONS (Cont'd)

form of the isotopes is of great significance because of the two orders of magnitude higher cross-section for the (alpha, neutron) reaction with F atoms compared to O atoms that are chemically bound to the alpha decaying nucleus.

Three different isotopic compositions were considered:

1. Weapons grade plutonium.

2. Dissolved fuel irradiated to 9% buildup of Pu-240.

3. Isotopic composition of smears from a deep bed Fiberglas filter (DBFF).

In addition, the oxide and fluoride chemical forms of TRU isotopes were used to estimate the TRU content of the tank contents. Transuranic (TRU) content based on different fractions of the plutonium content of the processed fuel being present in the waste, in addition to the fission products and transplutonium isotopes of a 9% Pu-240 burn up fuel, were also calculated.

The calculated Pu-239 values, with the TRU isotopes present as fluorides in the 11 ft layer of activity in Tank CX-72, are consistent with the measured bulk neutron production rate. The estimates of 51 and 0.65 kg of dissolved 9% Pu-240 burn up fuel are considered unrealistic because the waste discharged to the tank for terminal storage contained only the unrecovered plutonium that was originally present in the processed fuel. For this reason, a 1-2% loss of plutonium to the waste is necessary instead of 0.1% in the established and tested REDOX process. The estimated 150 to 200 g of plutonium in the tank corresponds to about 1% of unrecovered product present as fluoride in the waste discharged to the tank. This conclusion is not unreasonable because pilot scale studies were conducted in the initial stages of PUREX process development.

Additional support for the estimated 150 g of plutonium in tank CX-72 is provided by actual analyses of sludge specimens from tank CX-70. The reported 239/240-Pu activity was 253 uCi/L of centrifuged sludge that constituted 42 vol% of the specimen. This tank received waste exclusively from REDOX process flow sheet refinement investigations with <0.1% plutonium losses. Based on this result , the plutonium content of tank CX-72 was calculated as 12 g. Incorporation of 1% plutonium losses in the PUREX process (as compared to <0.1% in REDOX process), the plutonium content of tank 241-CX-72 is in excess of 120 g.

As a result of an objective assessment of pertinent information found in historical records and its technical evaluation, the fissile content of the 11 ft activity layer at the bottom of tank CX-72 can be concluded to range between 150 and 200 grams. This estimate requires confirmation by

4.0 DISCUSSION (Cont'd)

4.2 DISCUSSION OF RECENT RADIOLOGICAL INVESTIGATIONS (Cont'd)

actual analysis of a sample obtained from this region of the tank. A sampling of tank CX-72 contents, when implemented, can be augmented by analyzing the specimen for hazardous components.

The recorded absence in 1976 of any material in this tank during attempts at sampling and waste inspection could possibly be due to a compact, dry layer at the bottom of the tank with the remainder of the tank volume being void space. This interpretation is consistent with past documentation that the tank was empty, which led to the filling of the tank with grout in 1986. A soft structural integrity of the grout is indicated by the 2 ft core specimen retrieved recently from one of the 8-in. risers of the tank.

5.0 CONCLUSIONS

The following provides the radiological and chemical characterization legitimately considered to be probable for tank 241-CX-72.

Tank 241-CX-72 received waste from the PUREX process. The PUREX process used tributyl phosphate in kerosene solvent to extract plutonium and uranium from acid solutions of irradiated uranium. Nitric acid was used to promote extraction of plutonium and uranium. Process condensates from the PUREX process contained predominantly dilute nitric acid and other inorganic contaminants.

There is some indication through personal contacts with people once associated with the Strontium Semiworks that the decontamination flushes following the PUREX processes were also routed through Tank 241-CX-72. See Attachment 4 for a list of the decontamination solutions used.

No reliable historical radiological inventory estimates are available for Tank 241-CX-72. Tank 241-CX-72 was not directly associated with any other tanks. Speculation regarding the constituents of Tank 241-CX-72, based on known constituents of Tank 241-CX-70, is beyond the scope of this report. However, although the extraction efficiencies of the REDOX and the PUREX processes were different, the waste from processing irradiated material would be expected to contain the same species of radionuclides.

A formal waste designation has not been performed on the waste in this tank because the quantity, concentrations, and current chemical compositions are not known. The chemical composition of the chemicals used for the decontamination flushes is known (see Attachment 3), but the quantity used for each flush is not known.

Although it is uncertain whether the decontamination flushes went to Tank 241-CX-72, it is known that Sodium Dichromate was used in the decontamination flushes. If the decontamination flushes did indeed go to this tank, then the

5.0 CONCLUSIONS (Cont'd)

presence of chromium would cause the tank to be designated as a potentially DANGEROUS WASTE. Other constituents, especially fluoride, would also influence the DANGEROUS WASTE designation.

In addition to the above, the following conclusions are based on recent radiological investigations conducted by Dr. Vishnu B. Subrahmanyam of Analytical Systems Laboratories:

- There is strong evidence of the presence of a minimum of a 10 ft layer of radioactivity at the tank bottom.
- There is a step-wise increase (approximately ten fold) in radiation dose rates in 5-ft intervals.
- The inner walls of the tank are contaminated with Cs-137.
- The presence of essentially fast neutrons is confirmed.
- The activity layer is dry and contains little, if any, hydrogenous materials to thermalize the neutrons generated within the contents of the tank.
- The activity layer at the bottom of Tank 241-CX-72 contains fission product activities mixed with a most probable 150 to 200 grams of the fissile isotope, Pu-239.

The radiological and chemical constituents estimated for this tank can only be confirmed by direct analysis of representative specimens of the grout and sludge retrieved from the tank. This approach requires adherence to mandated sampling and documentation protocols.

6.0 ACKNOWLEDGEMENTS

It is a pleasure to acknowledge the support provided by Dr. Vishnu Subrahmanyam of Analytical Systems Laboratories. Dr. Subrahmanyam conducted recent radiological characterization investigations on Tank 241-CX-72 and worked diligently to provide the results of these investigations in time for inclusion in this report. His efforts on the recent radiological characterization investigations as well as his technical review of this report are greatly appreciated.

7.0 REFERENCES

The following documentation was reviewed to assist in the preparation of this report:

Westinghouse Document, SD-DD-TI-034, "Tank 241-CX-70 Waste Removal Assessment," October 31, 1988.

7.0 REFERENCES (Cont'd)

General Electric Document, HW-52860, "Standby Status Report Hot Semiworks Facility," September 1, 1957.

General Electric Document, HW-31767, "Hot Semiworks REDOX Study," January 27, 1954.

Rockwell Document, SD-DD-FL-001, "Rockwell Retired Contaminated Facility Listing and Description," July 19, 1982.

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Battelle Document, "Level 1 Remedial Investigation Work Plan - 200 Area Strontium Semiworks Liquid Waste Disposal Site," September 1987.

Rockwell Document, D0105ER0001, "Preliminary Study for Decontamination and Decommissioning of Strontium Semiworks," October 1, 1979.

Rockwell Document, SD-DD-ES-003, "Strontium Semiworks Decommissioning Engineering Study," July 1983.

Rockwell Document, RHO-HS-EV-28, "Criteria and Standards with Supporting Rationale for Decommissioning and Disposal of the Hot Semiworks Facilities," September 9, 1983.

Internal Letter, G. J. Pilicy to D. R. Speer, "Environmental Evaluation for Decommissioning Hot Semiworks," dated August 31, 1983.

Internal Letter, D. W. Fukumoto to W. M. Hayward, "Concrete Specifications for A, C, D, and E Cells, Galleries, Tanks, and Labyrinth," dated July 26, 1985.

Internal Letter, G. J. Carter, Jr. to D. R. Speer, "Concrete Fill of Hot Semiworks Facility," dated January 18, 1985.

Internal Letter, B. F. Weaver to W. F. Heine, "201-C Cell Tank Fill," dated February 24, 1986.

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Rockwell Document, RHO-CD-1541, "Hot Semiworks In Situ Disposal Study," September 1981.

General Electric Document, HW-72666, "Hot Semiworks Strontium-90 Recovery Program," July 17, 1963.

7.0 REFERENCES (Cont'd)

Report, "Effluent Controls Appraisal Report of Findings - Hot Semiworks/C Plant," February 7, 1981.

Rockwell Document, SD-DD-PP-001, "Strontium Semiworks Decommissioning Project Plan," August 1983.

Rockwell Document, SD-WM-SAR-003, "The Safety Analysis Report for the Decontamination and Decommissioning of Strontium Semiworks Complex," March 1984.

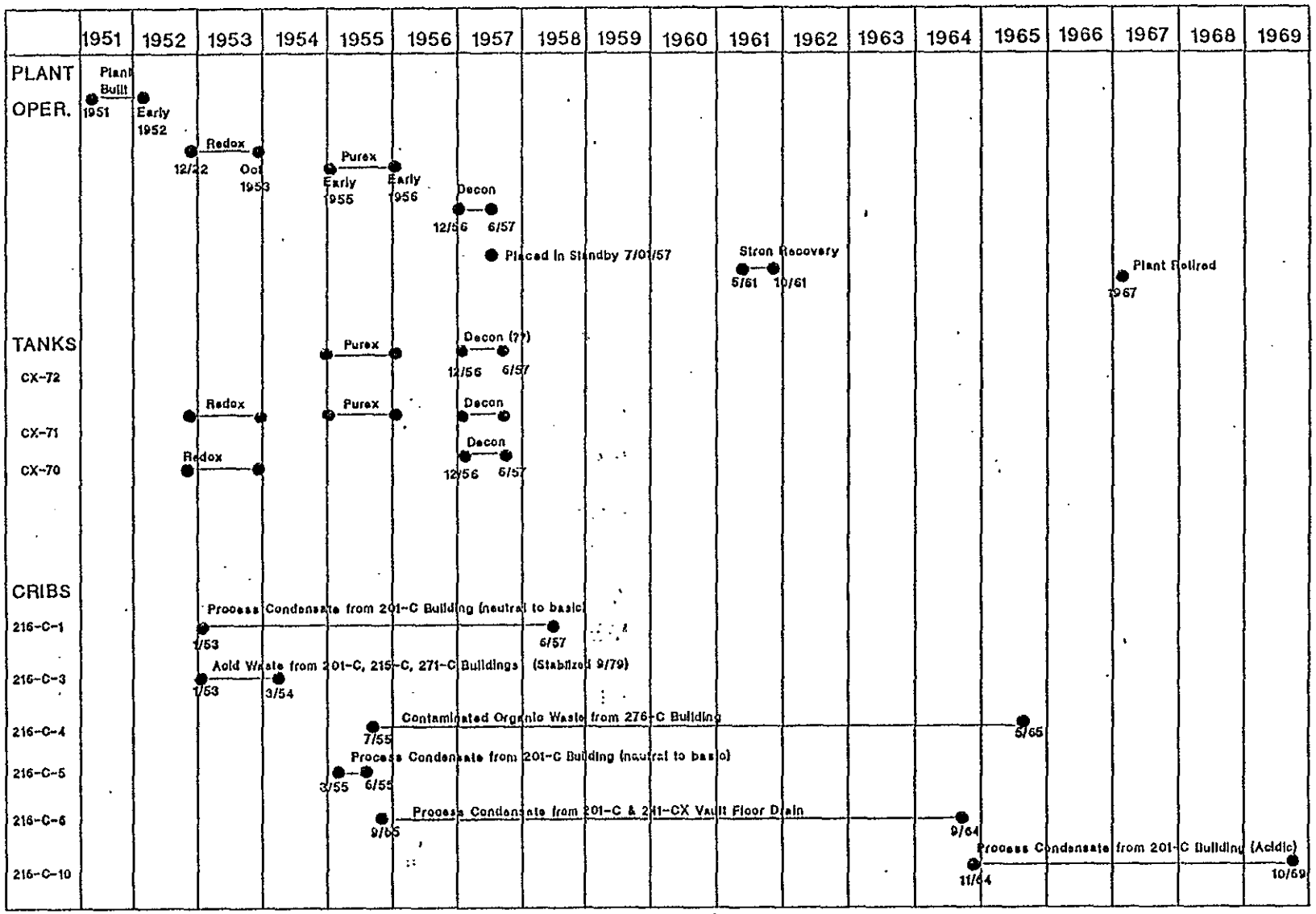
DOE Document, DOE/EA-0259, "Environmental Assessment Relating to the Decommissioning of Strontium Semiworks Facility," May 1985.

Letter, G. Burton to O. J. Elgert, "Waste Tank Survey," dated June 2, 1976.

8.0 ATTACHMENTS

ATTACHMENT 1

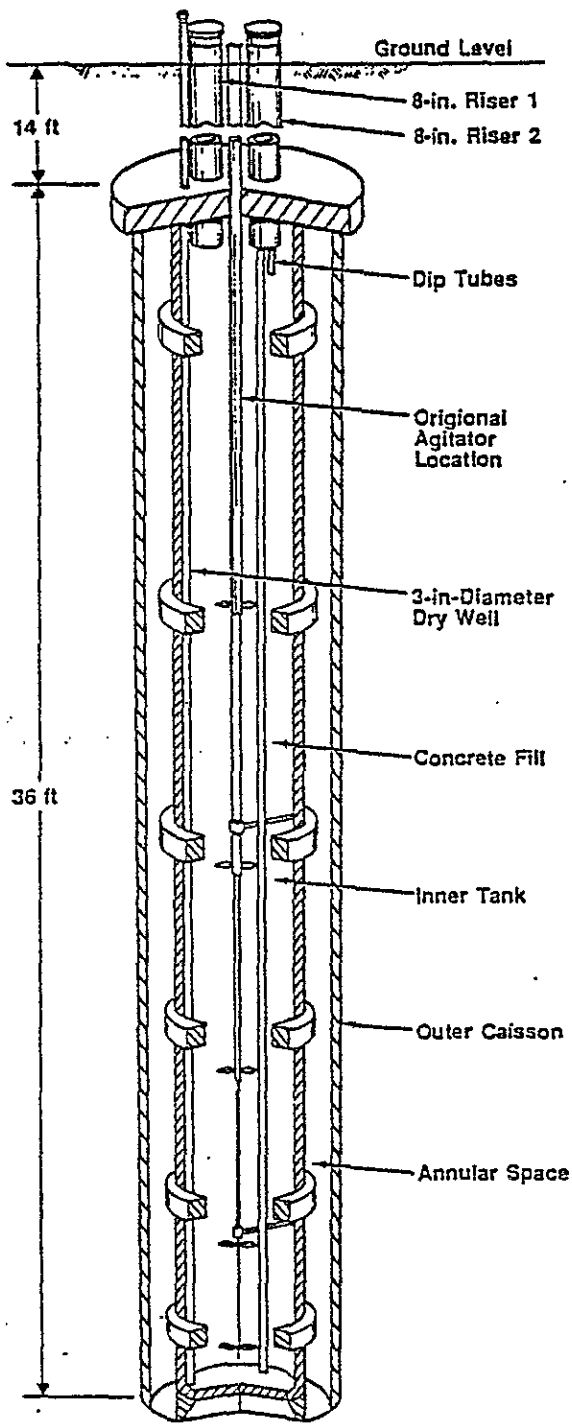
STRONTIUM SEMIWORKS TIME SEQUENCE



AUTHOR: J. E. Cummings 6/22/89

ATTACHMENT 2

TANK 241-CX-72 CONFIGURATION



ATTACHMENT 3

DECONTAMINATING SOLUTIONS USED FOLLOWING PUREX OPERATIONS

Contamination	Code	Composition (Wt. %)	Application Temperature
Ru	PC	1-1/2% $KMnO_4$ (Permanganate) 1/2% NaOH (Caustic)	60° C
Ru	CP	5% NaOH (Caustic) 2% H_2O_2 (Peroxide)	40° C
Zr-Nb	CT	6% NaOH (Caustic) 1-1/2% Tartaric Acid (Tartrate)	80-100° C
Zr-Nb	CTP	6% NaOH - 1-1/2% Tartaric acid - 2% H_2O_2 (Caustic-Tartrate-Peroxide)	25-30° C
Zr-Nb	HF	5% HNO_3 - 1% NaF (Nitric-Fluoride)	25-30° C
MnO_2 (from PC)	N-F-FAS	5% HNO_3 - 1% NaF - 2% Fe $(NH_4)_2$ $(SO_4)_2$ (Nitric-Fluoride-Ferrous Ammonium Sulfate)	25-30° C
MnO_2	N-FAS	5% HNO_3 - 2% Fe $(NH_4)_2$ $(SO_4)_2$ (Nitric-Ferrous Ammonium Sulfate)	25-30° C
All	3-20	6.9% NaF - 27.7% HNO_3 (3% HF - 20% HNO_3)	25-30° C
Zr-Nb	OX	5% $H_2C_2O_4$ (Oxalic Acid)	80° C
Grease plus FP	TSP	2% Na_3PO_4 (Trisodiumphosphate)	80-100° C
Ce, Cs	ND	6 Molar HNO_3 - 10% Sodium Dichromate	50° C
Pu	SD	Mixture of sulfuric acid and sodium dichromate	50° C
All	Oakite	10 - 20% Oakite #31	
on Masks	Turco	1 - 14% Turco 4182A	

NOTE: Exposure to solutions may vary from 15 minutes to 2 hours.

ATTACHMENT 4

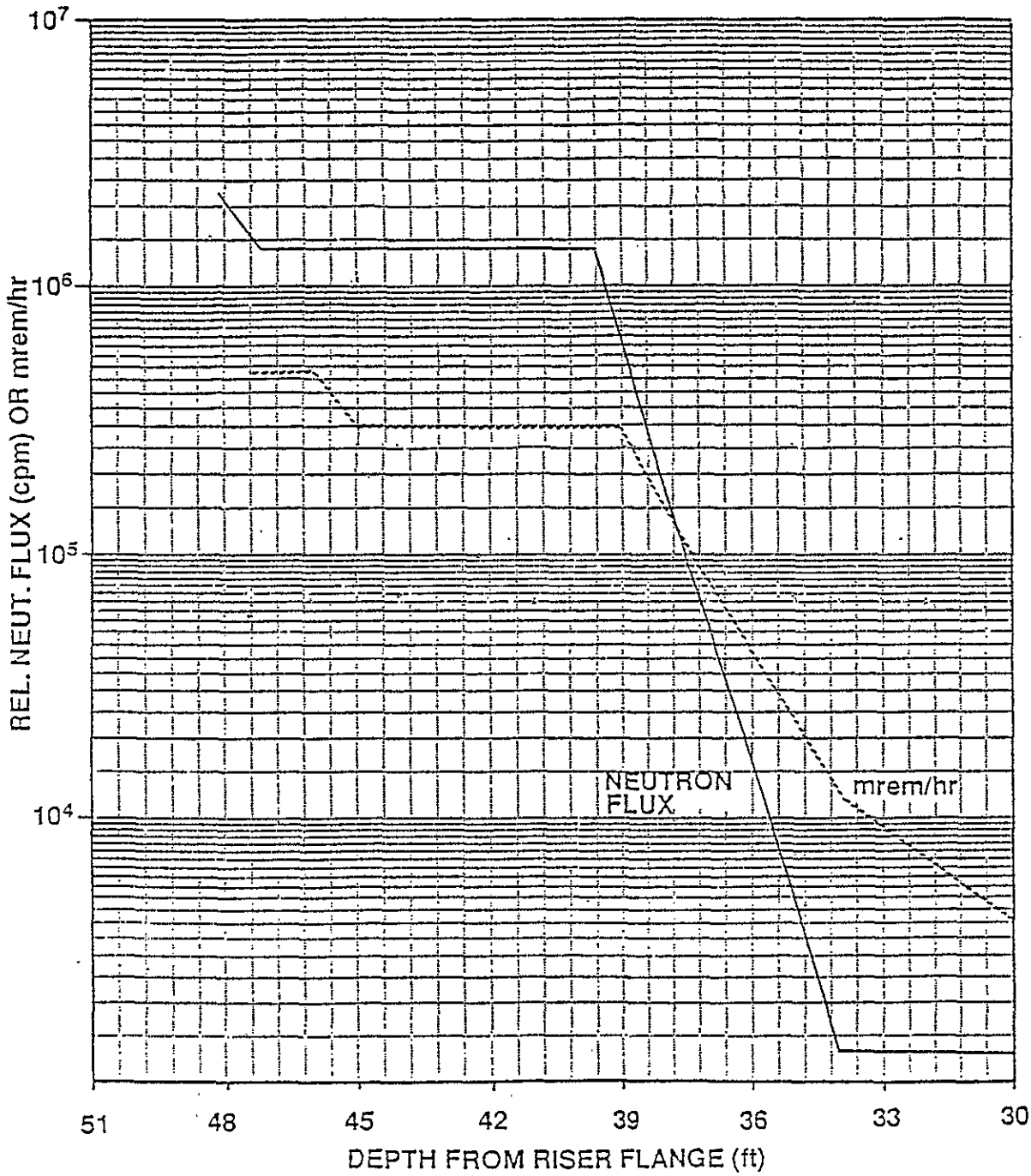
RADIONUCLIDE AND CHEMICAL COMPOSITION OF CX-70 SAMPLE

Component	Concentrations			
	Supernatant (M)	Slurry (M)	Water-Soluble[a]	Acid-Soluble[a]
SO_4^{-2}	0.061	0.059	3.1 E-05	*
CO_3^{-2}	0.033	**	2.3 E-05	**
TOC (gL)	0.312	0.598	3.0 E-04	4.8 E-04
Total alpha (uCi/L)	1.34	335	*	0.58
Total beta (uCi/L)	1.73 E-04	3.26 E-05	7.55	604
Cs-137 (uCi/L)	9.92 E-03	1.27 E-04	9.75	10.8
Sr-90 (uCi/L)	1.71 E-03	7.49 E-04	0.402	147
Am-241 (uCi/L)	*	*	*	*
Pu-239, 240 (uCi/L)	1.31	253	*	0.56
SpG	1.14	1.236	**	**

* Below detection limits.
** Insignificant amount.
a - Concentration as moles or microcuries per mL of centrifuged sludge.

ATTACHMENT 5

TANK 241-CX-72 - RELATIVE NEUTRON FLUX AND RADIATION DOSE PROFILES



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